

Sulphur budget in forested watersheds in Haute Ardenne region (Belgium)

P. Buldgen, J. Remacle

Ecologie microbienne, Département de Botanique, Université de Liège, Sart Tilman, B-4000 Liège, Belgique

ABSTRACT

The impact of acid precipitations is studied by comparing two watersheds, one is covered by caduceous species (Beech, Oak) and by Spruce, the other is covered by Spruce. The two watersheds also differ by the hydrogeology. The Spruce watershed exhibits more important subsurface runoff whereas the water retention time is much longer in the other one. The fluxes of H^+ , sulphates and other ions are observed in several compartments : precipitation, throughfall, seepage below organic layers, streamflow at the output of the watershed. The acidity of the water can be related to the anions, sulphate and nitrate. The chemistry of the streamflow and the buffering capacity of the watershed are controlled by biotic and abiotic factors : plant cover, microbial activities, hydrogeology.

INTRODUCTION

Sulphur compounds, especially sulphur dioxide and sulphate appear to be one of the most important component of acid rain.

Haute Ardenne region is covered by forests mainly by Spruce forests which are therefore the first target of acid rains. The impact of acid rains on forest ecosystems can be studied by several ways. In this work, it was decided to evaluate the biogeochemical fluxes. It needs an interdisciplinary approach (ecology, forestry, pedology, climatology, hydrogeology, microbiology) which leads to the knowledge of the hydrologic fluxes and the amounts of dissolved ions in a system which allows to draw up accurate budgets. It is the reason why forested watersheds were chosen. So it is possible to evaluate the inputs and outputs of the ecosystems without disturbing the ecological equilibrium. These data and the internal biochemical cycles are necessary to depict and quantify the transfer models within and through the forests. The chosen watersheds areas range in size from 80 to 90 ha. It allows to study representative samples of the Haute Ardenne region with a good insight of their characteristics. The watershed approach takes into account living as well as abiotic components and leads to quantitative and kinetics concepts. So the analysis of the transfers of acid rain through the forest upto the brook puts in light the influence of biotic and abiotic compartments and shows how an ecological system can respond to an environmental stress.

Table 1. Characteristics of the watersheds.

	Waroneu	Robinette
Area	83 ha	81 ha
Altitude	400 m	500 m
Slope	5-10 %	8 %
Precipitation	1200 mm	1270 mm
Streamflow	570 mm/year	560 mm/year
Underground water	4-29 mm	0-10 mm
Soils :		
pH		3.7 - 4.3
C/N		26 - 16
Clay		30 %
Silt		55 %
Sand		15 %
Plant cover (%)		
Spruce > 60 y. old	16	46
Spruce < 60 y. old	46	54
Beech	19	
Mixed caduceous forest	19	

The two forested watersheds are located in the hydrographic catchments of the rivers Gileppe and Soor at 500 m a.b.s. in Eastern Belgium. The "Robinette" watershed is covered by Spruce, 30 to 90 years old. The "Waroneu" watershed is covered by Spruce (60

%) and by caduceous trees (40 %) (Table 1).

The main characteristics of the watersheds are summarized in Table 1. Moreover, more specific observations were conducted in smaller forest stands : 0.5-3 ha covered either by spruce or by beech. The data collected in these stands will be used as bases for the ionic budget of both watersheds. The ionic concentrations and amounts were evaluated for four years in the following levels : precipitation, throughfall rain, seepage water below organic layers and the brook at the output of the watershed. More than 25,000 data were recorded and statistically analysed.

The paper summarized the main results which are fully discussed in several papers (see references).

RESULTS

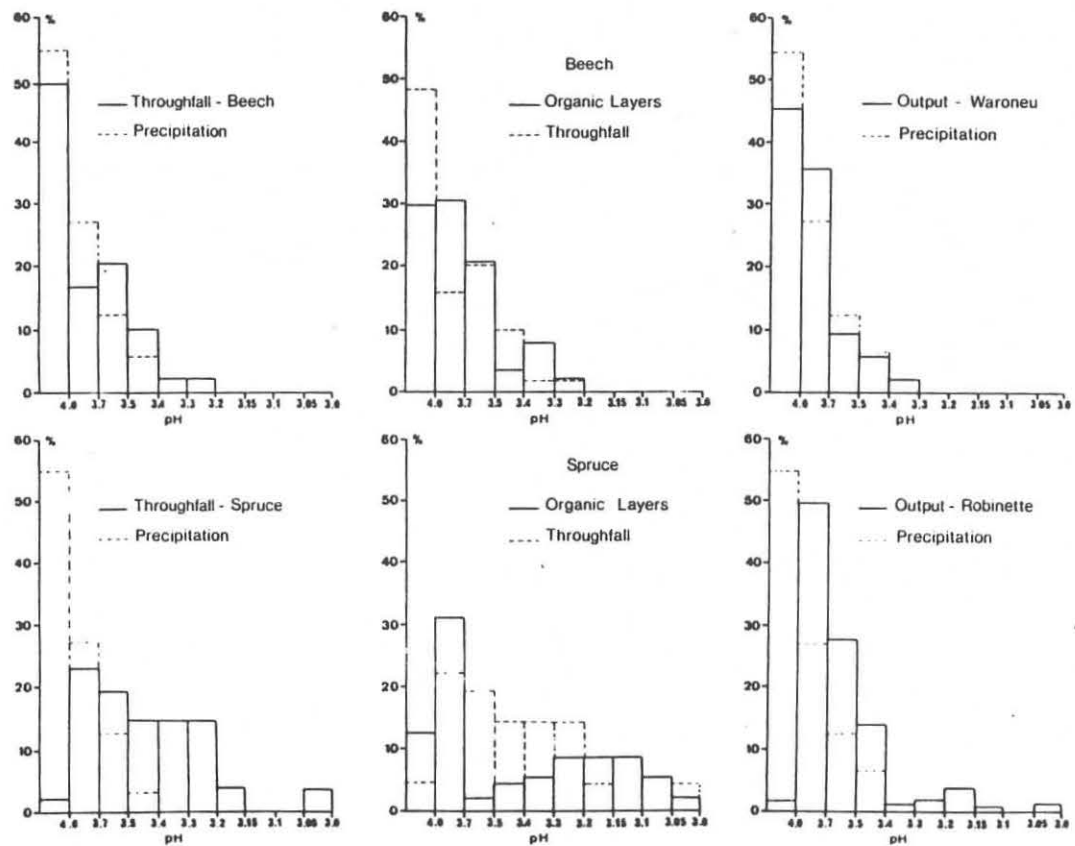
The following results will be commented :

- 1) The evolution of water acidity through both forested watersheds.
- 2) The relationship between acidity and the ionic composition of the water
- 3) The ionic concentrations of the water in the stream-water output of both watersheds
- 4) The ionic budgets for both watersheds

- 1) The evolution of the water acidity is followed through the forested watersheds by considering on the one hand the precipitation, the throughfall rain, the seepage water below the organic layers in two types of forest : spruce and beech forests, on the other hand the output of the two watershed i.e. the brooks Robinette and Waroneu. The stemflow was not taken into account because its weight is low in regard to the precipitation and throughfall. Figure 1 shows the distribution of monthly pH values observed during 4 years. More than 50 % of the pH values in the precipitation are below 4 units. pH values as low as 3.5 units have been recorded during some months. By flowing through the canopy, the rain becomes more acidic. It is specially true under Spruce where the frequency of pH lower than 3.5 units rises.

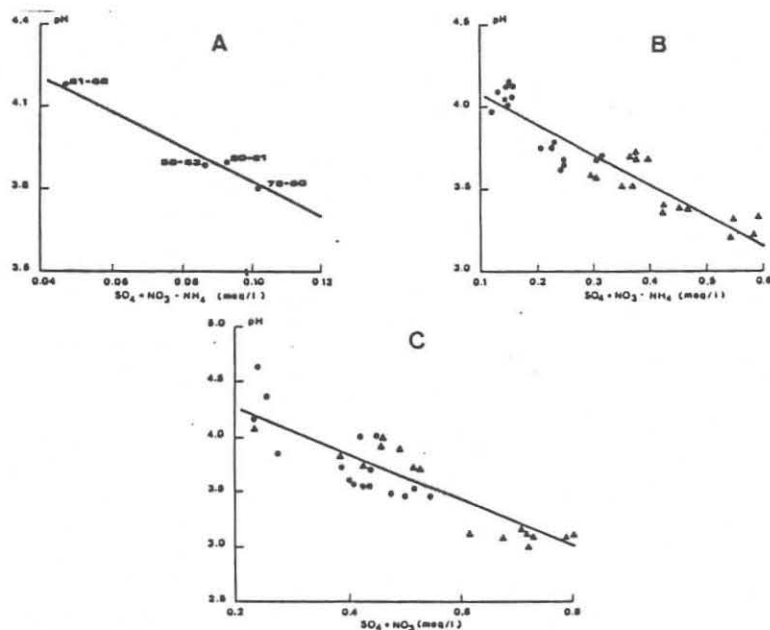
It can be also observed that the flow through the organic layers increases the acidity of the water mainly in spruce stands. The pH values peak around 3.3 and 3.1. However a second peak is noted between 3.7 and 4. It reflects that the values of two different spruce stands were analysed together. The water flowing through the mineral soil and the subsoil of the watersheds becomes less acid. It is obvious in the brook at the output of the watershed Waroneu. The same trend is observed, at a less extent, at the output of the watershed Robinette.

Figure 1. Distribution of pH values in precipitation, throughfall, seepage below the organic layers, streamflow (output of the watershed).



2) We shall now examine the relationships between the ionic composition and the acidity of the water (Fig. 2).

Figure 2. Relationship between acidity and ionic composition. A : in the precipitations; B : in the throughfalls; C : in the seepage below the organic layers (● Beech, ▲ Spruce).



The acidity of rainwater is well correlated with the relation : $(SO_4 + NO_3) - NH_4$. All concentrations are expressed as meq/l. It is clear that the acidity of the precipitations in Haute Ardenne depends on the gaseous emissions of sulphur and nitrogen compounds in the atmosphere.

The same correlation is observed for the acidity of throughfall rain under spruce and beech stands (Fig. 2) Moreover, it can be inferred that the throughfall rain is more acid below spruce than

beech because of a better trapping of nitrogen and sulphur oxides by spruce canopy. It results that the amounts of basic cations increase in throughfall. The question arises now to know whether acid rain promotes the foliar leaching of cations or whether basic cations are supplied by depositions with sulphates and nitrates. As it concerns potassium, our results show that the leaves and needles specially the old ones, release this cation. It is more difficult to assess this process for calcium, magnesium and sodium.

pH of seepage water below the organic layers is correlated with the sum of sulphate and nitrate anions (Fig. 2). Sulphates and nitrates are supplied in part by throughfall rains but microbial processes such as nitrification and sulfoxidation enhance the acidity. The influence of microbial activities upon the supply of nitrates and sulphates is proved by the relationship observed between the amounts of nitrates and sulphates in the seepage on one hand and the nature and thickness of the organic layers on the other hand. These latter parameters are obviously related to microbial activities. It must be added that the respiration of decomposers contribute also to the supply of H^+ in soil solution.

3. The mean annual ionic concentrations in the stream water output of the watersheds are summarized in Table 2.

Table 2. Annual average concentrations in stream-water output for both watersheds (mg/l).

	Waroneu	Robinette
H	0.133	0.253
Ca	1.12	0.70
Mg	1.72	1.10
Na	4.18	3.23
K	0.87	0.53
Fe	0.16	0.29
P- PO_4	0.013	0.013
N- NH_4	0.094	0.092
N- NO_3	1.60	2.47
S- SO_4	2.44	3.00
C	4.23	9.72

The main differences between the two brooks are :

- higher concentrations of calcium, magnesium, sodium and potassium in the brook Waroneu
- higher concentrations of H^+ , nitrates, sulphates, organic carbon and iron in the brook Robinette.

These differences can be mainly explained by the hydrogeological characteristics of both watersheds. The characteristics of the Waroneu watershed (Table 1) lead to a greater water retention time than in the Robinette watershed. For example, after a heavy rain resulting in a flood of 20,000 cubic meters, only 42 % of the rain flow in the stream of the Waroneu watershed instead of

73 % in the stream of the Robinette watershed within 3 days. It can be assumed that the loss of cations results, to a great extent, from the weathering of the rocks and exchange processes in the Waroneu watershed. In the Robinette watershed, the surface and subsurface run-off lead to high concentrations of organic carbon, iron, sulphate, nitrate and H^+ in the brook. As observed in the other compartments, the mobility of nitrates and sulphates anions is associated with a flux of H^+ . It appears thus that the impact of acid rains on the characteristics of the brooks depend on a lot of biotic and abiotic parameters.

- 4) The annual budgets of H^+ , sulphates and nitrates are given in Table 3. They are expressed as kg/ha.year and were drawn by taking into account the weight of the different forest covers (Table 1) and the rain interception by the canopies. The H^+ budget will be mainly discussed.

Table 3. Annual budgets of H^+ , sulphates and nitrates in the watersheds Waroneu and Robinette (kg/ha.year)

	WARONEU			ROBINETTE		
	H^+	$S \cdot SO_4$	$N \cdot NO_3$	H^+	$S \cdot SO_4$	$N \cdot NO_3$
1. Rain	1.4	18.4	10.3	1.5	19.5	11.0
2. Depositions	0.8	26.9	5.2	1.6	32.7	7.6
3. Throughfall	2.2	45.3	15.5	3.1	52.2	18.6
4. Seepage	2.3	44.5	16.3	3.7	53.2	21.0
5. Output	0.7	40.3	8.8	1.4	51.9	14.2
6. Uptake		8.1			9.7	
7. Immobilization		3.4			5.1	
8. Budget (1+2) - 5	+1.5	+5.0	+6.7	+1.7	+0.3	+4.4

As it concerns the Waroneu watershed, the supply of H^+ is due to, in a decreasing rank, precipitations, throughfall rains,

microbial processes in organic layers. In the Robinette watershed, the inputs of H^+ by precipitation and by dry-wet depositions are of the same order of magnitude because of the efficient trapping of depositions by spruces needles. The microbial processes contribute to a greater input of H^+ than in the Waroneu watershed. It must be mentioned that the cations uptake and root respiration are not considered. However, the expression :

$$H_{sto}^+ = H_{in}^+ - H_{ou}^+ \quad (1)$$

takes into account the internal H^+ production (microbial activities, cations uptake by roots, ...) and the H^+ consumption (anions uptake by roots, rocks weathering, ...),

H_{sto}^+ = H^+ storage in the watershed

H_{in}^+ = H^+ input (wet and dry depositions)

H_{ou}^+ = H^+ output in the brook

H_{sto}^+ equals 1.5 and 1.7 Kg/ha.year respectively in the watersheds Waroneu and Robinette. It accounts for 68 and 55 % of

H_{in}^+ . Besides, the output of H^+ can be also calculated by the following equation :

$$H_{ou}^+ = (H_{in}^+ + H_{pro}^+) - H_{neut}^+ \quad (2)$$

H_{pro}^+ : H^+ generated in the watershed

H_{neut}^+ : H^+ neutralized in the watershed.

By considering (1) and (2), it is clear that :

$$H_{neut}^+ = H_{sto}^+ + H_{pro}^+$$

By taking into account the data, it can be concluded that the amounts of H_{neut}^+ are of the same order of magnitude in both

watersheds. But the Robinette watershed is characterized by a rather poor buffer capacity owing to its geological and hydrological features. This infers that sulphate and aluminium outputs are more important in the Robinette watershed than in the Waroneu watershed. The Waroneu watershed stores, to some extent, the sulphate ions. In this watershed, the neutralization of H^+ leads to output of basic cations whereas the output of aluminium is not significant.

CONCLUSIONS

The results exemplify that the responses of forests against the acidic deposition depend on several features : biotic factors such as plant cover and microbial activities and abiotic factors such as the water retention time in the watershed.

These characteristics control the kinetic of ions transfers and therefore the soil and subsoil acidification and the losses of ions.

The knowledge of the homeostasis of forest ecosystems needs the study of control watersheds for several years.

This approach leads to a better understanding and quantification of the nutrients evolution and to accurate mathematical models of transfers.

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